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**FINAL TECHNICAL REPORT  
PREPARED FOR THE  
OFFICE OF NAVAL RESEARCH**

**TITLE: PRIMARY DISSOCIATION PROCESSES OF ENERGETIC COMPOUNDS**

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## ABSTRACT

During the last contract period, we have successfully constructed a very sophisticated and highly sensitive molecular beam apparatus for the investigation of primary thermal and photon induced dissociation processes of polyatomic molecules under isolated conditions. This machine incorporates many new ideas and has been shown to be the best molecular beam apparatus for this purpose in the world. It has enabled us to carry out a series of investigations which are relevant to the decomposition of energetic compounds. The understanding of the dynamics of concerted unimolecular decomposition, the determination of the energetics of important radical species and the elucidation of the primary dissociation mechanisms of various nitro and nitramine compounds are three areas in which we have made important contributions. (mgm) ←

The successful investigation of the primary dissociation of larger and less volatile molecules such as RDX was certainly the highlight of this contract research. Systematic investigations on concerted decompositions were carried out for a series of esters, ethers, nitrocompounds, and s-tetrazine. Through these investigations we have significantly furthered our understanding of the dynamics of the concerted decomposition of various organic molecules.

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Toward the end of the contract period, we have also constructed a picosecond laser system for the study/investigation of intramolecular energy transfer processes and a fourth generation crossed molecular beams apparatus.

**I. Brief Summary of Scientific Accomplishments for ONR Contract No. N00014-83-K-0069 entitled "Primary Dissociation Processes of Energetic Compounds."**

**(1) Construction of a State-of-the-Art Molecular Beam Translational Spectrometer.**

Molecular beam photofragmentation translational spectroscopy is one of the best methods for investigating primary thermal and photon induced dissociation processes in polyatomic molecules. Whether concerted reactions form stable molecular products with a large release of translational energy (which ensemble averages to a boom) is an important question in the study of energetic materials, and can be answered unambiguously with this technique.

Because the original molecular beam machines were designed to do crossed molecular beams experiments, they are not optimized for crossed laser beam-molecular beam experiments. During the contract period, in order to improve the situation, we developed a new state-of-the-art molecular beam machine, specifically for molecular photofragmentation translational spectrometry, which incorporates many new ideas and is designed to give higher resolution, higher sensitivity and lower background than the molecular beam apparatus which existed

previously. The construction of the new machine was completed recently. A series of experiments carried out for the purpose of evaluating various aspects of its performance clearly indicates that this apparatus is working as well as expected from our design criteria. With this apparatus, which is by far the best in the world, we have started the long awaited investigation of primary dissociation processes of molecules of energetic materials. A schematic diagram is shown in Fig. 1. The instrument consists of three parts: a rotating molecular beam source, a fixed, ultra-high vacuum, mass-spectrometric detector, and a "main" interaction chamber.

A continuous molecular beam is formed by expanding a reagent through a 0.125 mm nozzle into the source region. The source region is pumped by the 6" diffusion pumps which do not move with the rotating source and maintain a pressure of  $10^{-4}$  torr when the beam is on. The beam is skimmed and a region of differential pumping intervenes between the source region and the main chamber. The differential region is normally at  $10^{-5}$  torr and is pumped by a 350 liter/sec turbomolecular pump which rotates with the source. A pulsed laser beam, which propagates along the rotation axis of the beam source, crosses the molecular beam creating a pulse of dissociation products.

A small angular fraction of this pulse of products will travel through two regions of differential pumping, spreading as it goes due to the velocity distribution of the nascent

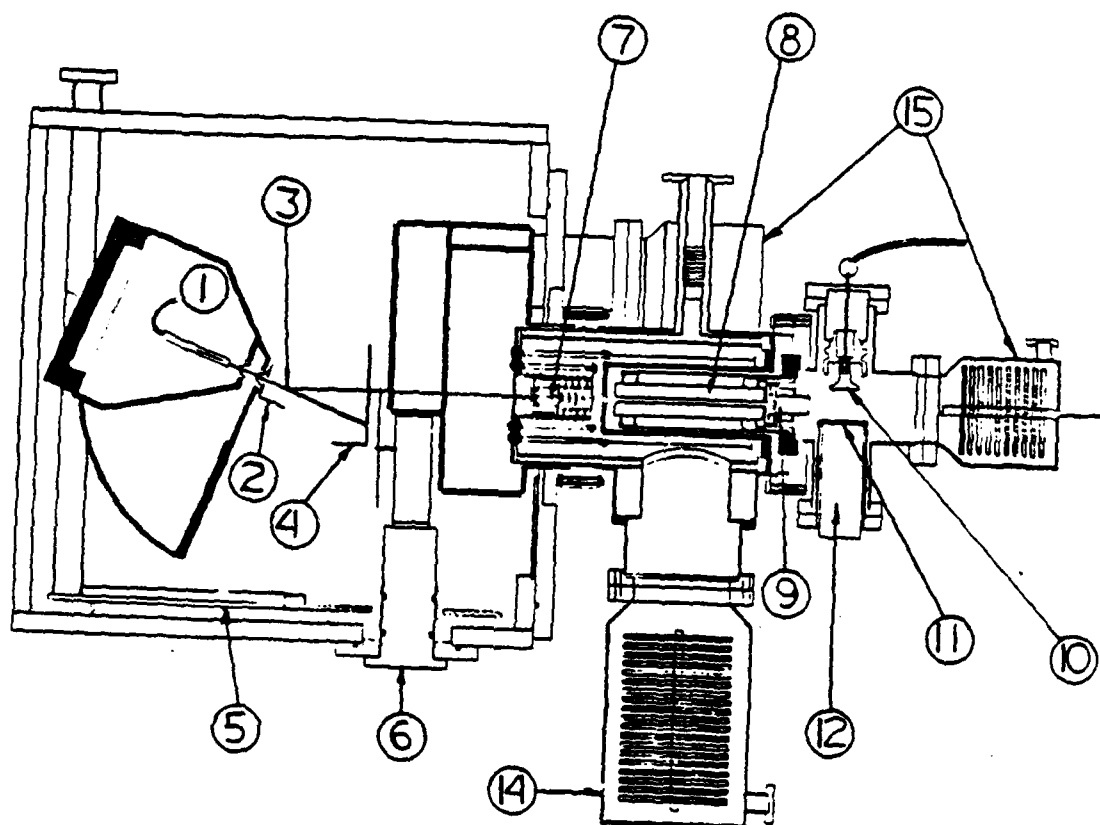


Fig. 1. The new molecular beam photodissociation machine. (1) Gas feed line to nozzle. (2) Liquid helium cooled collimation slit. (3) Interaction zone. (4) Liquid nitrogen cooled beam catcher. (5) Liquid nitrogen cooled copper panels. (6) Removeable time-of-flight wheel, for measuring primary beam velocity distribution. (7) Brink-type ionizer. (8) Quadrupole mass filter. (9) Exit ion optics. (10) Ion target. (11) Scintillator. (12) Photomultiplier tube. (14) Magnetically suspended turbomolecular pump. (15) Grease sealed turbomolecular pumps.

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products. The pulse of dissociation products is then ionized in a liquid nitrogen cooled chamber (typically  $10^{-10}$  torr), by a Brink-type, electron-impact ionizer. Ions products are directed through a quadrupole mass filter to a Daly-type ion counter, made up of an ion target, a scintillator and a photomultiplier tube. The transient pulse or time-of-flight (TOF) spectrum is recorded on a 1024 channel scaler operating as a pulse counter with a minimum channel width of 1  $\mu$ sec, which is triggered by the laser pulse. For high TOF resolution using a 1  $\mu$ sec channel width, a digital delay can be introduced on the trigger of the multi-channel scaler so that the transient pulse is recorded in as many of the 1024 channels as possible. The data acquisition is overseen and directed by a macro program running on a DEC LSI-11 lab computer.

The holes in the detector walls between 3 and 7 in Fig. 1 are precision machined and define the "viewing window" of the detector. These are placed so that the detector "sees" all parts of the 3mm x 3mm x 1mm interaction volume (at 3) for all angles to which the beam source can rotate. The most effective way of reducing the background pressure in the detector in a dynamic vacuum system is by the installation of differential pumping. However, for those molecules that travel straight through all of the differential pumping apertures, this so-called "direct-through background" cannot be decreased by any number of differential pumping regions. At a pressure of  $2 \times 10^{-7}$  torr in the main chamber, the mean free path of the

molecule is much longer than the dimensions of the chamber itself. For this reason "direct through background" cannot come from collisions with other molecules within the viewing window of the detector. Consequently, a closed cycle refrigerator is used to cool a copper collimation slit to 30°K at 2 in Fig. 1. This lowers the background originating from the main chamber by a factor of ten. In addition to this, liquid nitrogen is supplied to the main chamber and cools large copper panels which serve as cryopumps, creating an effective beam catcher for condensible gases.

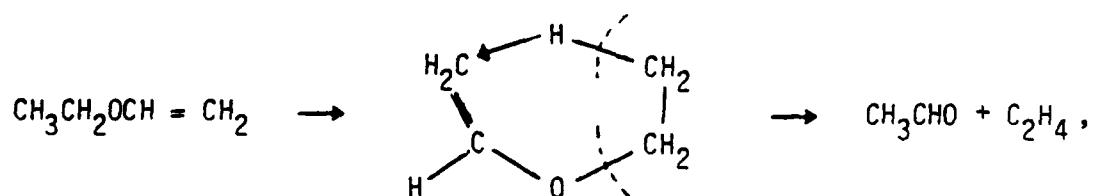
(2) **Dynamics of Concerted Unimolecular Decomposition of Ethyl Vinyl Ether and Diethyl Ether.**

Most energetic compounds are characterized by having a large potential energy barrier which separates the reactant from fragment products which are much lower in energy. The decomposition of these molecules can proceed either through simple bond rupture, producing radicals, or through a concerted reaction, producing molecular products. Since decomposition to radical products is endothermic, chemical energy of energetic materials can only be released in the subsequent radical chain reactions. On the other hand, concerted reactions forming molecular products promptly release the available chemical energy in the primary step.

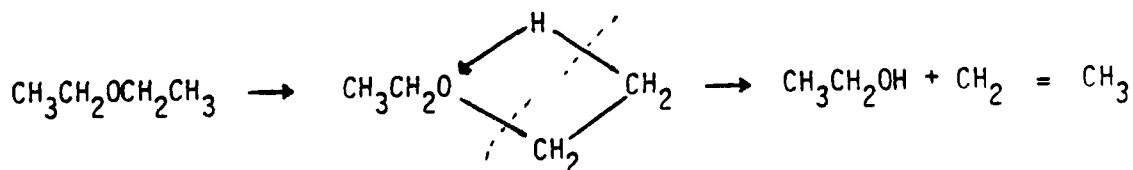
There is special interest on the dynamics of energy disposal in a concerted dissociation. Whether the chemical energy is released into vibrational-rotational excitation or

translational energy might have significant consequences in the macroscopic behavior of energetic materials.

The first important observation made on this topic, carried out under the ONR contract, revealed very surprising results. In the decomposition of ethyl vinyl ether, in which a hydrogen atom from the ethyl group is transferred to the end carbon atom of the vinyl group through a six membered cyclic transition state,



a large fraction of the potential energy of the exit barrier is released as translational energy. The astonishingly high mean translational energy of 31 kcal/mol indicates that the molecule dissociates "explosively" after electron rearrangement takes place. The dissociation of diethyl ether, in which a hydrogen atom from the end carbon transfers to the oxygen atom through a four membered cyclic transition state



shows similar behavior.

- (3) **Photodissociation of Acetylene at 193.3 nm.** J. Phys. Chem. 89, 4744 (1985).

We have measured the translational energy release for processes  $C_2H_2 \xrightarrow{193\text{ nm}} C_2H + H$  (I) and  $C_2H \xrightarrow{193\text{ nm}} C_2 + H$  (II) using the molecular beam time-of-flight method. By measuring the maximum release of translational energy for process I it has been determined that the C-H bond energy in acetylene,  $D_0(C_2H-H)$ , is  $132 \pm 2$  kcal/mol. Since the translational energy distribution peaks well away from zero, it is unlikely that internal conversion of the electronically excited  $C_2H_2$  is an important channel for dissociation in channel I. Because of the relatively small amount of energy appearing as product rotation, it is possible to get information on the vibrational structure of the  $C_2H$  radical from the translational energy distribution and it is found that the bending frequency in  $C_2H$  is  $550 \pm 100\text{ cm}^{-1}$ . Our experiment is consistent with earlier work which observed  $C_2(^1\pi_u)$  in fluorescence.

- (4) **The Observation of  $CH_3O$  in the Collision-Free Multiphoton Dissociation of  $CH_3NO_2$ .** J. Chem Phys. 84, 1044 (1986).

The method of infrared laser, crossed molecular beam photofragmentation translational spectroscopy has been used to study the "thermal" chemistry of nitromethane under collision-free conditions. We have observed for the first time experimental evidence that shows that the isomerization of nitromethane to methylnitrite is an important thermochemical process in the unimolecular decomposition of the title

compound. Based on theoretical predictions of Dewar et al. that the Arrhenius pre-exponential factor is  $10^{13.3}$ , we have been able to estimate the potential energy barrier height for the isomerization reaction to be  $55.5 \pm 1.5$  kcal/mol, compared to the C-N bond energy of 59.2 kcal/mol. It is clear that in mechanistic modeling of the thermal decomposition of nitromethane, this important dissociation channel must be taken into consideration. We have also been able to conclusively show that previous suggestions of a dynamical barrier in the exit channel of the simple C-N bond fission reaction are in error.

(5) **Infrared Multiphoton Dissociation of Three Nitroalkanes.**  
J. Phys. Chem. 90, 3549 (1986).

Infrared multiphoton dissociation in a molecular beam has been studied in order to elucidate the collision-free "thermal" chemistry and dynamics of nitromethane, nitroethane, and 2-nitropropane. The isomerization of  $\text{CH}_3\text{NO}_2$  to  $\text{CH}_3\text{ONO}$  was observed by detecting the  $\text{CH}_3\text{O}$  and NO products from the dissociation of the internally very hot, isomerized nitromethane. A novel application of RRKM theory was used to estimate the barrier height to isomerization at 55.5 kcal/mol. The barrier height determination method was tested and found to give excellent results by applying it to the determination of the barrier height of HONO elimination from nitroethane, a value which is well-known from activation energy measurements. The method was then applied to the case of HONO elimination

from 2-nitropropane and it appears that there is good reason to believe that the barrier height is 3-5 kcal/mol lower in 2-nitropropane than in nitroethane. The success of this method for determining barrier heights shows how a "microscopic" molecular beam experiment, using infrared multiphoton dissociation, where the concept of temperature has no place, can be quantitatively related to pyrolysis experiments which are conducted under collisional, thermal conditions and measure phenomenological quantities such as activation energies. The concerted HONO elimination reactions from nitroethane and 2-nitropropane were found to channel about 70 and 65% of the exit barrier into translational energy, respectively. This large release of translational energy is suggested to be due to the nature of the transition-state mechanical barrier which is largely made up of repulsive energy between the closed shell products and not of reactant strain energy. The small difference between nitroethane and 2-nitropropane in the translational energy distributions is explained in terms of a scaled reduced mass impulse approximation that is used to characterize the repulsive excitation dynamics of the departing closed shell products.

- (6) **Infrared Multiphoton Dissociation of RDX in a Molecular Beam.** J. Chem. Phys. **88**, 801 (1988).

Infrared multiphoton dissociation (IRMPD) of hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) in a molecular beam has been performed in order to investigate the mechanism of RDX thermal decomposition. A beam of molecules was crossed by a pulsed TEA CO<sub>2</sub> laser and velocity distributions of the various products were measured by the time-of-flight (TOF) technique as a function of laboratory angle using a mass spectrometric detector. The dissociation channels, their branching ratios, and the translational energy distributions of the products were determined. In contrast to the conventional view of simple bond rupture through loss of NO<sub>2</sub> as the dominant primary channel in RDX decomposition, it was found that the dominant primary channel is concerted symmetric triple bond fission to produce three CH<sub>2</sub>N<sub>2</sub>O<sub>2</sub> fragments which subsequently undergo secondary concerted dissociation to produce HCN, H<sub>2</sub>CO, HONO (or HNO<sub>2</sub>), and N<sub>2</sub>O. A total of two primary and four secondary dissociation channels were observed. Concerted reactions predominate over simple bond rupture not only in the number of channels (four versus two) but also in the amount of products. A fair amount of translational energy release through concerted reaction channels was observed, which is significant for an explanation of the energies of RDX decomposition.

- (7) **Infrared Multiphoton Dissociation of Ethyl and Methyl Acetate.** J. Phys. Chem. **92**, 5379 (1988).

The collisionless decomposition of ethyl and methyl acetate was investigated with IR multiphoton dissociation. With a fluence of  $40 \text{ J/cm}^2$ , ethyl acetate gave 97% concerted decomposition producing ethylene and acetic acid, the latter of which underwent significant secondary decomposition to ketene and water. Simple bond rupture producing ethyl radical and  $\text{CH}_3\text{CO}_2$ , which completely decomposed to  $\text{CH}_3$  and  $\text{CO}_2$ , accounted for the remaining reaction products. Methyl acetate underwent a concerted reaction to produce methanol and ketene, and simple bond rupture to form  $\text{CH}_3$  and  $\text{CH}_3\text{CO}_2$ , in near equal amounts. All of the concerted reactions released more than half ( $\sim 20$  kcal/mol) of the exit channel barrier into translational energy. From the branching ratio between the two channels and the translational energy distribution of the simple bond rupture channel, the barrier for concerted reaction in methyl acetate was determined to be  $69 \pm 3$  kcal/mol.

- (8) **A Concerted Triple Dissociation-The Photochemistry of s-Tetrazine.** J. Chem. Phys. (in press) (1989).

The method of photofragment translational spectroscopy was used in a molecular beam study of the photochemistry of s-tetrazine. Following  $^1\text{B}_{3u} \leftarrow ^1\text{A}_g$  ( $\text{S}_2 \leftarrow \text{S}_0$  excitation) or  $^1\text{B}_{2u} \leftarrow ^1\text{A}_g$  (248 nm excitation), s-tetrazine reverts to the highly vibrationally excited ground electronic state through internal conversion, then decomposes into  $2\text{HCN} + \text{N}_2$  via

concerted triple dissociation. By analyzing the experimental data with a simple model, it was found that the potential energy surface along the reaction coordinate after the transition state is very repulsive, leading to most of the available energy, on average 73.9% in the case of  $S_1 \leftarrow S_0$  excitation, going into translation. The asymptotic angles between  $N_2$  and HCN with respect to the center-of-mass of s-tetrazine were  $117.2 \pm 0.5^\circ$  and  $114.4 \pm 0.5^\circ$  for  $S_1 \leftarrow S_0$  and 248 nm excitation, respectively.

**(9) Construction of a Picosecond Laser System for the Study of Intramolecular Energy Transfer Processes.**

In order to study intramolecular energy transfer processes, a picosecond laser system has been constructed. This laser system uses a continuous-wave mode-locked Nd:YAG laser as the oscillator. The  $1.06 \mu\text{m}$  pulses generated from the oscillator are first compressed and then frequency doubled to  $0.53 \mu\text{m}$ . These green pulses are then used to synchronously pump two dye lasers to obtain two independent picosecond pulses which are tunable from 570 to 700 nm. These pulses are further amplified by a chain of dye cells pumped by a frequency doubled Nd:YAG laser operated at 10 Hz. One of the tunable visible pulses is first doubled to the UV then frequency tripled in a rare gas cell with a LiF window to produce VUV pulses for resonant photoionization detection.

The construction of this laser system was delayed partly because of the move of our laboratory from Lawrence Berkeley Laboratory to Giauque Hall at the University of California, Berkeley in 1986, and partly because of the frequent failure of the continuous-wave mode-locked Nd:YAG laser. A new laser was acquired to replace the old laser and the system is now ready to go.

**(10) Construction of a Fourth Generation Crossed Molecular Beams Apparatus.**

In order to be able to study many important chemical reactions involving radical molecules, we have undertaken the construction of a fourth generation crossed molecular beams apparatus.

This new apparatus will use extensive cryopumping in both the main chamber and the detector chamber in order to reduce the background noise level significantly. In the detector chamber, the ultrahigh vacuum pumping arrangement has to be made in such a way that both rare gases and hydrocarbons can be removed efficiently. The combined use of newly developed magnetically suspended turbomolecular pumps and liquid helium cryopumping seem to offer the best solution.

A high power Koch closed-cycle refrigeration system with a cooling power of 200 W at 20°K and 60W at 4°K has been acquired. After careful design and fabrication of the cryogenic components, we are almost ready for the conversion of

an old apparatus into a fourth generation machine. This conversion is expected to be completed some time in the fall.

**List of Publications of ONR Supported Research**

1. Photodissociation of Acetylene at 193.3 nm, Alec M. Wodtke and Y.T. Lee, J. Phys. Chem. **89**, 4744 (1985).
2. The Observation of  $\text{CH}_3\text{O}$  in the Collision Free Multiphoton Dissociation of  $\text{CH}_3\text{NO}_2$ , Alec M. Wodtke, Eric J. Hints, and Yuan T. Lee, J. Chem. Phys. **84**, 1044 (1986).
3. Infrared Multiphoton Dissociation of Three Nitroalkanes, A. M. Wodtke, E.J. Hints, and Y.T. Lee, J. Phys. Chem. **90**, 3549 (1986).
4. Molecular Beam Studies of Elementary Chemical Processes, Yuan T. Lee, Nobel Lecture, 1986.
5. High-Resolution Photofragmentation-Translational Spectroscopy, in Advances in Gas-Phase Photochemistry and Kinetics, Molecular Photodissociation Dynamics, edited by M.N.R. Ashfold and J.E. Baggott, Royal Society of Chemistry (1987) pp.31-59.
6. Infrared Multiphoton Dissociation of RDX in a Molecular Beam, Xinsheng Zhao, Eric J. Hints, and Yuan T. Lee, J. Chem. Phys. **88**, 801 (1988).
7. Infrared Multiphoton Dissociation of Ethyl and Methyl Acetate, Eric J. Hints, Alec M. Wodtke, and Yuan T. Lee, J. Phys. Chem. **92**, 5379 (1988).
8. A Concerted Triple Dissociation - The Photochemistry of S-Tetrazine, Xinsheng Zhao, Walter B. Miller, Eric J. Hints, and Yuan T. Lee, J. Chem. Phys. (submitted) (1988).

9. Photodissociation of Cyclic Compounds in a Molecular Beam, Xinsheng Zhao, Ph.D. thesis, University of California, Berkeley, 1988.
10. Molecular Beam Photodissociation Studies of Polyatomic Molecules and Radicals, E.J. Hintsa, Ph.D. thesis, University of California, Berkeley, 1989.

#### Invited Lectures Presented During the Contract Period

1. Yuan T. Lee, Molecular Beam Studies of Primary Photochemical Processes of Organic Molecules, Department of Chemistry, University of Utah, Salt Lake City, Utah, December 7, 1982.
2. Yuan T. Lee, Molecular Beam Chemical Kinetics, Golden Jubilee of Chinese Chemical Society Located in Taipei, Taipei, Taiwan, China, December 12, 1982.
3. Y.T. Lee, Primary Photochemical Processes by Molecular Beam Photofragmentation Translational Spectroscopy, International Conference on Photochemistry and Photobiology, January 3-8, 1983, Alexandria, Egypt.
4. Y.T. Lee, Crossed Molecular Beams Studies Using the Seeded Supersonic Beams Method, John Fenn Symposium, Yale University, March 14-1, 1983.
5. Y.T. Lee, Dynamic Resonances in Reactive Scattering, Department of Chemistry, Rice University, March 16, 1983.
6. Y.T. Lee, Elucidation of Reaction Mechanisms by the Crossed Molecular Beams Method, Kolthoff Lecture, Department of Chemistry, University of Minnesota, April 25, 1983.
7. Y.T. Lee, Molecular Beam Studies of Primary Photochemical Processes, Kolthoff Lecture, Department of Chemistry, University of Minnesota, April 27, 1983.
8. Y.T. Lee, Dynamic Resonances in the  $F + H_2$  Reaction, Kolthoff Lecture, Department of Chemistry,<sup>2</sup> University of Minnesota, April 29, 1983.

9. Y.T. Lee, High Brilliance VUV Photochemistry, Workshop on Synchrotron Radiation Source, Lawrence Berkeley Laboratory, May 10, 1983.
10. Y.T. Lee, The Effect of Internal Excitation in Promoting a Chemical Reaction, Workshop on Elementary Chemical Reactions, University of Paris, Orsay, France, June 10, 1983.
11. Y.T. Lee, Recent Advances in Molecular Beam Chemistry, Workshop on Elementary Chemical Reactions, University of Paris, Orsay, France, June 10, 1983.
12. Y.T. Lee, Reactive Scattering, Advances and Perspectives, International Symposium on Molecular Beams, Freiburg, West Germany, June 13-17, 1983.
13. Y.T. Lee, Introductory Remark on Reactive Scattering, Conference on Dynamics of Molecular Collisions, Gull Lake, Minnesota, June 27-July 1, 1983.
14. Y.T. Lee, Molecular Beam Studies of Primary Photodissociation Processes, XI International Conference on Photochemistry, University of Maryland, College Park, Maryland, August 21-26, 1983.
15. Y.T. Lee, UV Photodissociation of  $1,2-C_2F_4BrI$ : Bond Selective Photodissociation through Electron Transition, Herrsching Workshop on Primary Photophysical Processes, Herrsching, West Germany, October 16-21, 1983.
16. Y.T. Lee, Molecular Beam Studies of Primary Photochemical Processes of Polyatomic Molecules, Department of Chemistry, California Institute of Technology, Pasadena, California, October 28, 1983.
17. Y.T. Lee, Elucidation of Reaction Mechanisms Involving Oxygen Atoms with Unsaturated Hydrocarbons, FMC Lecture, Department of Chemistry, Princeton University, Princeton, New Jersey, November 2, 1983.
18. Y.T. Lee, Experimental Investigations of Resonance Phenomena in Elementary Chemical Reactions, FMC Lecture, Department of Chemistry, Princeton University, Princeton, New Jersey, November 2, 1983.
19. Y.T. Lee, Photodissociation of Alkyl Polyhalides Using an Excimer Laser, FMC Lecture, Department of Chemistry, Princeton University, Princeton, New Jersey, November 2, 1983.

20. Y.T. Lee, Reactions of Oxygen Atoms with Unsaturated Hydrocarbons, Frontier Lecture, Texas A&M, College Station, Texas, November 27, 1983.
21. Y.T. Lee, Promotion of Chemical Reaction by Reagent Translation and Internal Excitation, Frontier Lecture, Texas A&M, College Station, Texas, November 28, 1983.
22. Y.T. Lee, Experimental Probing of the Transition State in the  $F + H_2$  Reaction, Frontier Lecture, Texas A&M, College Station, Texas, November 29, 1983.
23. Y.T. Lee, Infrared Multiphoton Dissociation of Polyatomic Molecules, Frontier Lecture, Texas A&M, College Station, Texas, November 30, 1983.
24. Y.T. Lee, Molecular Beam Photofragmentation Translational Spectroscopy, Frontier Lecture, Texas A&M, College Station, Texas, December 1, 1983.
25. Y.T. Lee, Dynamics in Mechanisms of Primary Photochemical Processes of Polyatomic Molecules, Department of Chemistry, University of Rochester, Rochester, New York, December 4, 1983.
26. Y.T. Lee, Molecular Beam Studies of Elementary Chemical Reactions, Harrison Howe Award Lecture, Rochester Section of the ACS, Rochester, New York, December 4, 1983.
27. Y.T. Lee, Direct Probing of the Transition State: Dynamic Resonances in the  $F + H_2$  Reaction, Department of Chemistry, California Institute of Technology, Pasadena, California, December 8, 1983.
28. Y.T. Lee, Formation and Decay of Quasi-bound States in Elementary Chemical Reactions, International Conference on Radiationless Transitions, Newport Beach, California, January 2-7, 1984.
29. Y.T. Lee, Molecular Beam Studies of Elementary Chemical Reactions, University of Ohio, Columbus, Ohio, February 29, 1984.
30. Y.T. Lee, Photofragmentation Translational Spectroscopy, University of Ohio, Columbus, Ohio, March 1, 1984.
31. Y.T. Lee, Dynamic Resonances in the Reaction of Fluorine Atoms with Hydrogen Molecules, 187th American Chemical Society Meeting, St. Louis, Missouri, April 9-12, 1984.

32. Y.T. Lee, Molecular Beam Studies of Reaction Dynamics, University of Wisconsin, Madison, Wisconsin, April 18, 1984.
33. Y.T. Lee, Energetics of Radicals Involved in Combustion Processes, Department of Energy Combustion Science Contractor's Meeting, Sandia National Laboratory, Livermore, California, May 22-23, 1984.
34. Y.T. Lee, Dynamics and Mechanisms of Elementary Chemical Reactions, Symposium on "Frontiers of Chemistry," American Association for Advancement of Science, New York, New York, May 24-27, 1984.
35. Y.T. Lee, Dynamic Resonances in the  $F + H_2$  Reaction, Fifth American Conference on Theoretical Chemistry, Jackson, Wyoming, June 15-20, 1984.
36. Y.T. Lee, Dynamics and Mechanisms of Elementary Chemical Reactions, 188th American Chemical Society National Meeting, Philadelphia, Pennsylvania, August 26-28, 1984.
37. Y.T. Lee, Direct Probing of the Transition State: Dynamic Resonances in the  $F + H_2$  Reaction, The Fifth European Conference on Dynamics of Molecular Collisions, Jerusalem, Israel, September 3-7, 1984.
38. Y.T. Lee, Energetics and Dynamics of Chemical Reactions by Crossed Molecular Beams Experiments, University of Pittsburgh, Pittsburgh, Pennsylvania, September 13, 1984.
39. Y.T. Lee, Dynamic Resonances in the Reaction of Fluorine Atoms with Hydrogen Molecules, Stanford University, Stanford, California, October 11, 1984.
40. Y.T. Lee, Molecular Beam Chemistry, 31st G.N. Lewis Memorial Lecture, University of California, Berkeley, California, October 23, 1984.
41. Y.T. Lee, Primary Dissociation Processes of Polyatomic Molecules, ONR/NRL Workshop on Energetic Materials Initiation Fundamentals, Baltimore, Maryland, October 30-31, 1984.
42. Y.T. Lee, Dynamic Resonances in Reactive Scattering, University of Illinois, Champaign-Urbana, Illinois, November 7, 1984.
43. Y.T. Lee, Molecular Beam Studies of Primary Photochemical Processes, Lasers' 1984, San Francisco, California, November 29, 1984.

44. Y.T. Lee, Crossed Molecular Beam Studies of Dynamics of Chemical Reactions, Academia Sinica, Taipei, Taiwan, December 17, 1984.
45. Y.T. Lee, State Specificity in the Reaction of Electronically Excited Sodium Atoms with Oxygen Molecules, Xth International Symposium on Molecular Beams, Cannes, France, June 2-7, 1985.
46. Y.T. Lee, Recent Advances in Reactive Scattering, International Conference on Recent Advances in Molecular Reaction Dynamics, Aussois, France, June 9-14, 1985.
47. Y.T. Lee, Molecular Beam Studies of Reaction Dynamics, Freie Universitat, West Berlin, Germany, June 18, 1985.
48. Y.T. Lee, Dynamics and Kinetics of Elementary Chemical Reactions, National Taiwan University, Taipei, Taiwan, August 2, 1985.
49. Y.T. Lee, Molecular Beam Photofragmentation Translational Spectroscopy on  $C_2H_2$  using 193 nm Photons, XI International Photochemistry Conference, Tokyo, Japan, August 4-9, 1985.
50. Y.T. Lee, Reaction of Electronically Excited Sodium Atoms with Simple Molecules, Fudan University, Shanghai, China, August 10, 1985.
51. Y.T. Lee, Dynamics of Concerted Unimolecular Decomposition Through Cyclic Transition States, Institute of Chemistry, Beijing, China, August 13, 1985.
52. Y.T. Lee, Resonances and Orbital Alignment in Elementary Chemical Reactions, Institute of Chemistry, Beijing, China, August 14, 1985.
53. Y.T. Lee, State Specificity and Orbital Alignment Dependence in the Reaction of Electronically Excited Sodium Atoms, Institute of Chemical Physics, Dalian, China, August 16, 1985.
54. Y.T. Lee, Bond Selective Photochemistry on  $CH_2BrI$  Through Electronic Transitions, Institute of Chemical<sup>2</sup> Physics, Dalian, China, August 17, 1985.
55. Y.T. Lee, Vibrational Predissociation Spectroscopy of Protonated Hydrogen Clusters, 1985 American Chemical Society Meeting, Symposium on "Spectroscopy and Dynamics of Highly Vibrationally and Rotationally Excited Polyatomic Molecules," Chicago, Illinois, September 9-13, 1985.

56. Y.T. Lee, The Heat of Formation of  $C_2H$  and  $C_2H_3$  Radicals from Molecular Beam Photodissociation, 1985 American Chemical Society Meeting, Symposium on "Transient Species," Chicago, Illinois, September 9-13, 1985.
57. Y.T. Lee, Recent Investigations of Reaction Dynamics, Photochemistry and Spectroscopy by the Molecular Beam Method, Institute for Molecular Science, Okazaki, Japan, March 20, 1986.
58. Y.T. Lee, Energetics and Dynamics of the Concerted Unimolecular Decomposition of Organic Molecules, Institute for Molecular Science, Okazaki, Japan, April 9, 1986.
59. Y.T. Lee, Reaction Dynamics of Electronically Excited Alkali Atoms with Simple Molecules, Award Lecture, Debye and Langmuir Award Symposium, 191st ACS Meeting, New York, April 14, 1985.
60. Y.T. Lee, Current Status and Future Prospects of Chemical Kinetics, 1st National Meeting on Chemical Kinetics, Donxi, China, May 15, 1986.
61. Y.T. Lee, Experimental Probing of Critical Regions of Potential Energy Surfaces, Symposium on Recent Progress in Chemical Reaction Dynamics Research, Institute for Molecular Science, Okazaki, Japan, May 23, 1986.
62. Y.T. Lee, Investigation of Reaction Dynamics from Product Angular Distributions, Symposium on "Recent Progress in Chemical Reaction Dynamics Research," Institute for Molecular Science, Okazaki, Japan, May 24, 1986.
63. Y.T. Lee, Crossed Molecular Beam Studies of Reaction Dynamics, Department of Chemistry, Tohoku University, Sendai, Japan, May 26, 1986.
64. Y.T. Lee, IR Multiphoton Decomposition of Nitrocompounds, Tokyo University, Tokyo, Japan, May 28, 1986.
65. Y.T. Lee, Molecular Beam Studies of Electronically Excited Atoms, Tokyo University, Tokyo, Japan, May 29, 1986.
66. Y.T. Lee, Recent Investigations of Reaction Dynamics, Spectroscopy, and Photochemistry by the Molecular Beam Method, Department of Chemistry, Keio University, Yokohama, Japan, May 30, 1986.
67. Y.T. Lee, Understanding Chemical Reactions through Molecular Collisions, Keio University, Yokohama, Japan, May 31, 1986.

68. Y.T. Lee, Reaction Dynamics of Electronically Excited Sodium Atoms with Oxygen and Hydrogen Halides, Kyushia University, Fukuoka, Japan, June 2, 1986.
69. Y.T. Lee, Molecular Beam Studies of Primary Photochemical Processes, Department of Chemistry, Kyoto University, Kyoto, Japan, June 10, 1986.
70. Y.T. Lee, Infrared Absorption Spectroscopy of Ionic Clusters Using the Ion Trapping Technique, Department of Molecular Engineering, Kyoto University, Kyoto, Japan, June 11, 1986.
71. Y.T. Lee, Dynamics and Energetics of Primary Photochemical Processes, Plenary Lecture, XIII Informal Photochemistry Conference, Boulder, Colorado, June 23, 1986.
72. Y.T. Lee, A New Experimental Method for IR Absorption Spectroscopy on Ionic Clusters, Atomic and Molecular Science Institute, Academia Sinica, Taipei, Taiwan, July 25, 1986.
73. Y.T. Lee, Molecular Beam Studies on the C-H Overtone Excitation in Benzene, American Chemical Society Annual Meeting, Anaheim, California, September 8, 1986.
74. Y.T. Lee, Recent Advances in Molecular Beam Studies of Reaction Dynamics and Spectroscopy, Chemical Physics Retreat, University of Oregon, Eugene, Oregon, September 27, 1986.
75. Y.T. Lee, Primary Decomposition Processes of RDX, Workshop on Energetic Materials Initiation Fundamentals, Los Alamos, New Mexico, October 14, 1986.
76. Y.T. Lee, Spectroscopy and Dynamics of Laser and Molecular Beams, Regional Meeting on Chemical Physics, University of Waterloo, Waterloo, Ontario, Canada, October 25, 1986.
77. Y.T. Lee, Molecular Beam Chemical Kinetics, Department of Chemistry, University of California, Riverside, California, November 10, 1986.
78. Y.T. Lee, Molecular Beam Studies of Elementary Chemical Processes, Nobel Lecture, Royal Swedish Academy, Stockholm, Sweden, December 8, 1986.
79. Y.T. Lee, Infrared Absorption Spectroscopy of Hydrated Hydronium Ions by the Ion Trapping Technique, Department of Chemistry, University of Stockholm, Stockholm, Sweden, December 9, 1986.

80. Y.T. Lee, Mode Selective and Bond Selective Photochemical Processes, Chemical Society of Upsalla, Upsalla, Sweden, December 12, 1986.
81. Y.T. Lee, Concerted Unimolecular Decomposition of Organic Compounds, University of Gothenburg, Sweden, December 15, 1986.
82. Y.T. Lee, Crossed Molecular Beam Investigations of Chemical Reaction Dynamics, National Taiwan University, Taipei, Taiwan, December 18, 1986.
83. Y.T. Lee, Homogeneous Linewidth of C-H Overtone Excitation in Benzene Molecules, Annual Meeting of the Chinese Chemical Society, Tsinghua University, Hsinchu, Taiwan, China, December 19, 1986.
84. Y. T. Lee, Dynamics of Molecular Collisions, Lawrence Berkeley Laboratory, Physics Division, January 22, 1987.
85. Y. T. Lee, Infrared Absorption Spectroscopy of Hydrated Hydronium Ions, Chemistry Department, University of Southern California, Los Angeles, California, February 25, 1987.
86. Y. T. Lee, Energetics of Radical Molecules, Sandia National Laboratory Combustion Facility, Livermore, California, March 13, 1987.
87. Y. T. Lee, Crossed Molecular Beam Studies of Elementary Atomic and Molecular Processes, Stanford Linear Accelerator Center, Stanford, California, March 16, 1987.
88. Y. T. Lee, Dynamics of Elementary Chemical Reactions, Arthur Davidson Lecture, Department of Chemistry, University of Kansas, Kansas City, Kansas, March 24, 1987.
89. Y. T. Lee, Molecular Beam Studies of Chemical Reaction Dynamics, Department of Chemistry, Arizona State University, Tempe, Arizona, March 27, 1987.
90. Y. T. Lee, Molecular Beam Studies of Dynamics of Chemical Reactions, Sir Run Run Shaw Lecture, State University of New York, Stony Brook, New York, April 22, 1987.
91. Y. T. Lee, Infrared Absorption Spectroscopy of Ionic Clusters by the Ion Trapping Technique, Committee on Atomic and Molecular Science (CAMS) Workshop, National Research Council, Washington, DC. April 27, 1987.

92. Y. T. Lee, Molecular Beam Studies of Photochemical Processes, Plenary Lecture, International Conference on Quantum Electronics, Baltimore, Maryland, April 29, 1987.
93. Y. T. Lee, Dynamics of Elementary Chemical Reactions, Chinese Academy of Sciences, Beijing, China, May 13, 1987.
94. Y. T. Lee, Laser Chemistry and Laser Spectroscopy, Institute of Chemistry, Academia Sinica, Beijing, China, May 18, 1987.
95. Y. T. Lee, Crossed Molecular Beam Studies of Elementary Chemical Reactions, Nankai University, Nanking University, Fudan University, Institute of Chemical Physics, China, May 16-26, 1987.
96. Y. T. Lee, Molecular Beam Studies of Hot Atom Chemical Reactions, Plenary Lecture, XIII International Hot Atom Chemistry Symposium, Mt. Fuji, Japan, May 24-29, 1987.
97. Y. T. Lee, Two Way Street Between Dynamics and Spectroscopy, Plenary Lecture, Symposium on Reaction Dynamics, University of Tokyo, Tokyo, Japan, June 3, 1987.
98. Y. T. Lee, Reactions of Electronically Excited Sodium Atoms, 1987 Conference on Dynamics of Molecular Collisions, Oglebay Park, Wheeling, West Virginia, July 13, 1987.
99. Y. T. Lee, Molecular Beam Studies of the Primary Photodissociation Processes of Polyatomic Molecules, Plenary Lecture, XIIIth International Conference on Photochemistry, Budapest, Hungary, August 10, 1987.
100. Y. T. Lee, Crossed Molecular Beams Studies of Elementary Chemical Reactions, Selman A. Waksman Lecturer, October 7, 1987.
101. Y. T. Lee, Molecular Beam Studies of Elementary Chemical Reactions, (A) Nobel Lecture Series, Clark Kerr Campus, University of California, Berkeley, October 13, 1987; (B) Aerospace Corporation, Los Angeles, California, October 22, 1987; and (C) Convocation Lecture, MacAlester College, Minneapolis, Minnesota, October 28, 1987.
102. Y. T. Lee, Deriving Correct Conclusions from Experimental Observations, Department of Chemistry, MacAlester College, Minneapolis, Minnesota, October 29, 1987.
103. Y. T. Lee, Molecular Dissociations and Molecular Spectroscopy using Lasers and Molecular Beams, MacAlester College, Minneapolis, Minnesota, October 30, 1987.

104. Y. T. Lee, Molecular Beam Studies of Photophysical and Photochemical Processes, Plenary Lecture, International Laser Symposium-III, Atlantic City, New Jersey, November 2, 1987.
105. Y. T. Lee, Dynamics of Molecular Dissociations and Photochemical Processes by the Molecular Beams Method, Distinguished Lecture Series, Office of Naval Research, Washington, D.C., November 3, 1987.
106. Y. T. Lee, Investigation of Reaction Dynamics by the Molecular Beams Method, Plenary Lecture, Annual Meeting of the American Vacuum Society, Anaheim, California, November 4, 1987.
107. Y. T. Lee, Disposal of Angular Momentum in Elementary Chemical Reactions, Laser Chemistry Symposium, Kenting, Taiwan, China, November 12, 1987.
108. Y. T. Lee, Dependence of Molecular Orientation and Orbital Alignment in the Reactions of Electronically Excited Sodium Atoms, Laser Chemistry Workshop, Kenting, Taiwan, China, November 13, 1987.
109. Y. T. Lee, Unimolecular dissociation of the RDX Molecule by Molecular Beam Photofragmentation Translational Spectroscopy, Laser Chemistry Workshop, Kenting, Taiwan, China, November 14, 1987.
110. Y.T. Lee, Reactions of Electronically Excited Atoms, Memorial Symposium on the 100th Birthday of Otto Stern, University of Hamburg, Hamburg, West Germany, February 4, 1988.
111. Y.T. Lee, New Approach for IR Absorption Spectroscopy of Ionic Clusters, USA-USSR Linear and Non-Linear Laser Workshop, University of California, Santa Barbara, California, March 3, 1988.
112. Y.T. Lee, Elementary Chemical Reactions Involved in Combustion, University of Gothenburg, Sweden, March 21, 1988.
113. Y.T. Lee, Molecular Beam Studies of Chemical Reaction Dynamics, Department of Chemistry, University of Florida, Gainesville, Florida, March 28, 1988.
114. Y.T. Lee, Application of Molecular Beam and Laser Techniques in the Investigation of Spectroscopy and Photochemistry, Department of Chemistry, University of Florida, Gainesville, Florida, March 30, 1988.

115. Y.T. Lee, Crossed Molecular Beams Studies of Elementary Chemical Reactions, Mack Memorial Award Lecture, Department of Chemistry, Ohio State University, Columbus, Ohio, March 31, 1988.
116. Y.T. Lee, Bond Selective Photodissociation Through Electronic Transitions, Department of Chemistry, Ohio State University, Columbus, Ohio, April 1, 1988.
117. Y.T. Lee, Molecular Beam Studies on Atomic and Molecular Processes, Department of Physics, University of California, Berkeley, California, April 13, 1988.
118. Y.T. Lee, Molecular Beam Studies of Elementary Chemical Reactions, IBM Research Laboratory, San Jose, California, April 15, 1988.
119. Y.T. Lee, Molecular Collisions and Dynamics of Elementary Chemical Reactions, I to VIII, Department of Chemistry, Nanjing University, Nanjing, China, May 16-20, 1988.
120. Y.T. Lee, Endothermic Substitution Reactions of Aromatic Hydrocarbons, 2nd National Conference on Chemical Kinetics, Beijing, China, May 23, 1988.
121. Y.T. Lee, Elementary Chemical Reactions Involved in Combustion Chemistry, DOE Combustion Contractor's Meeting, Lake Geneva, Wisconsin, June 3, 1988.
122. Y.T. Lee, Molecular Beam Studies of Hot Atom Chemical Reactions, 3rd Chemical Congress of North America, Toronto, Canada, June 7, 1988.
123. Y.T. Lee, A New Approach for High Resolution Vibrational Spectroscopy on Solvated Ionic Species in the Gas Phase, 43rd Molecular Spectroscopy Symposium, Ohio State University, Columbus, Ohio, June 15, 1988.
124. Y.T. Lee, Chemistry in Modern Society, University of Metropolitana, Caracas, Venezuela, June 27, 1988.
125. Y.T. Lee, Application of Molecular Beam Techniques in the Investigation of Elementary Chemical Reactions, 1st Venezuela National Congress of Chemical Industry, Caracas, Venezuela, June 28, 1988.

126. Y.T. Lee, Substitution Reactions of Br Atoms with o-, m-, and p-chlorotoluene, Institute of Atomic and Molecular Sciences, Taipei, Taiwan, July 8, 1988.
127. Y.T. Lee, "Recent Advances in Chemical Reaction Dynamics, Department of Chemistry, University of Hong Kong, July 11, 1988.
128. Y.T. Lee, Infrared Absorption Spectroscopy of Hydrated Hydronium Ions, Department of Chemistry, University of Hong Kong, July 13, 1988.
129. Y.T. Lee, Molecular Beam Chemistry, Naval Research Laboratory, Washington, DC, August 5, 1988.
130. Y.T. Lee, Unimolecular Decomposition of Benzene, Thermochemistry and Kinetics, Current Directions Symposium, University of Southern California, September 22, 1988.
131. Y.T. Lee, Molecular Beam Chemical Kinetics, Department of Chemistry, Xiamen University, Xiamen, China, October 12, 1988.
132. Y.T. Lee, Investigation of Laser Induced Chemical Processes by the Molecular Beam Technique, Institute for the Structure of Matter, Chinese Academy of Sciences, Fuzou, China, October 14, 1988.
133. Y.T. Lee, Primary Photochemical Processes by Lasers and Molecular Beams, Department of Chemistry, University of Singapore, Singapore, October 21, 1988.
134. Y.T. Lee, New Spectroscopy Method for the Investigation of Solvated Hydronium and Ammonium Ions, Chemistry Department, National University Singapore, October 21, 1988.
135. Y.T. Lee, Molecular Collisions, Scientific Research, and Modern Society, Singapore, October 22, 1988.
136. Y.T. Lee, Molecular Beam Studies on Elementary Chemical Reactions, Haworth Lecture, Department of Chemistry, Brookhaven National Laboratory, Upton, Long Island, New York, November 15, 1988.
137. Y.T. Lee, IR Absorption Spectroscopy of Solvated Hydronium Ions, Department of Chemistry, Brookhaven National Laboratory, Upton, Long Island, New York, November 17, 1988.

138. Y.T. Lee, Molecular Beam Studies on Primary Photochemical Processes, Department of Physics, Kansas State University, Manhattan, Kansas, December 1, 1988.
139. Y.T. Lee, Dynamics on Elementary Chemical Reactions, Cardwell Lecture, Department of Physics and Chemistry, Kansas State University, Manhattan, Kansas, December 1, 1988.
140. Y.T. Lee, Molecular Collisions and Chemical Reactions, Ramon Areces Foundation, Madrid, Spain, December 15, 1988.
141. Y.T. Lee, Crossed Molecular Beams Studies of Elementary Chemical Reactions, University of Salamanca, Salamanca, Spain, December 16, 1988.